Pressure Effect on the Spin Fluctuations in YMn_2

–Mn NQR Study–

K. Nishikido[†], C. Thessieu[†], G-q. Zheng[†], Y. Kitaoka[†], K. Asayama[†], R. Hauser[‡]

[†] *Dpt of Mat. Physics, Osaka University, Osaka 560*

[‡] *Institut für Experimentalphysik, a-1040 Wien, Austria*

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Abstract

We report the NQR study on the pressure-induced paramagnetic state in the antiferromagnetic (AF) intermetallic compound YMn_2 with the Néel temperature T_N=100 K at ambient pressure. From the T variation of the nuclear spin-lattice relaxation rate, $^{55}(1/T_1)$ of ^{55}Mn above the critical pressure of 4 kbar, the spin fluctuation feature is found to change below a temperature T_{sf}. At higher temperature than T_{sf}, the nuclear relaxation behavior is well described in terms of the self-consistent renomalized (SCR) spin fluctuation theory for nearly AF metals, whereas below T_{sf} a deviation is significant and $1/T_1T$ exhibits a weak T variation. It is pointed out that T_{sf} coincides with the temperature below which a T^2 law in electrical resistivity is valid as expected for a Fermi liquid ground state. We proposed that these features are understood from the standpoint that the development of AF spin fluctuation remains in short-range associated with the singlet formation among Mn spins in each tetrahedron as suggested by the inelastic neutron experiments on $Y_{1-x}Sc_xMn_2$.

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I. INTRODUCTION

In a series of intermetallic compound RMn_2 (R=Rare Earth), an existence of magnetic moments on Mn sites depends largely on an inter-atomic Mn-Mn distance[1]. Above a critical distance of $d_{\text{Mn-Mn}}$=2.7 10^{-10}m there exists a magnetic moment on the Mn which is antiferromagnetically ordered in general. Below this critical distance the ground state remains paramagnetic down to lowest temperature. This fact that the Mn-Mn inter-atomic distance plays a key-role in determining their magnetic properties indicates that these compounds may be magnetically sensitive to such a perturbation as a chemical substitution or an application of pressure. An interesting possibility is then that an application of pressure induces Quantum Critical Phase Transition governed by the zero point quantum spin fluctuation. In particular this is the case of YMn_2 which exhibits antiferromagnetic (AF) order with a Néel temperature around 100 K at ambient pressure. A relatively low pressure of $P_{\rm c}$=3 to 4 kbar [1] is enough to suppress the AF order[2]. The paramagnetic to AF magnetic transition upon cooling is of first order type accompanying by well pronounced hysteretic phenomena in various measurements. Therefore, the AF to paramagnetic phase transition across P_c is anticipated to be first order type. So far it is reported that the paramagnetic state at ambient pressure is dominated by the AF spin fluctuations[3] which is well treated in terms of the self-consistent renormalized (SCR) spin fluctuation theory developed by Moriya et al.[4]. As a matter of fact, the Sc-substitution of Y suppresses the AF order and the spin fluctuations in the paramagnetic phase was reported to be well described in terms of the SCR theory[5]. On the other hand, the inelastic neutron scattering experiment has found that the singlet correlation among four Mn spins forming the tetrahedron is developed to stabilize this paramagnetic phase, i.e., a kind of spin liquid state is formed in the Sc substituted system. Thus, it is still controversial whether or not this novel character of spin fluctuations is actually related to the nature in the paramagnetic state above $T_{\rm N}$ in YMn_2 because the

[1]The value of the critical pressure depends on the experimental technique.

effect of disorder induces by the Sc -substitution is not addressed yet. In other words, we address the question whether or not, near the critical point in YMn_2 where the Néel temperature is suppressed to zero, the AF spin fluctuations are also critical with a damping energy tending to zero. An application of pressure to suppress the AF order is promising to settle these issues, because this is the only way to stabilize the paramagnetic state in YMn_2 at low temperatures without any disorderliness. The aim of this paper is to enligthten the evolution of low-energy magnetic excitations in the pressure-induced paramagnetic phase by approaching the critical point from the higher pressure side.

II. EXPERIMENTAL DETAILS

A polycrystalline sample has been crushed in fine powder in order to ensure the penetration of the radio-frequency pulsed field. A conventional clamp-type pressure cell was employed for the NMR experiment under pressure with use of the Fluorinert liquid as the pressure transmitting medium. The Fluorinert volume was of the proportion as the sample volume and froze below 200 K. To calibrate the pressure at the sample position at low temperature, the pressure shift in T_c of lead powder was monitored. The measurement of T_c was performed by an inductive method using the in-situ NMR coil in a resonant circuit. The NQR spectrum has been obtained by plotting the spin-echo intensity as a function of frequency with an interval of 50 kHz. The spin-lattice relaxation time, T_1, was measured by the saturation recovery method in a temperature range of $4.2 - 120$ K. For the NQR transition of $\pm 5/2 \leftrightarrow \pm 3/2$, the recovery function of nuclear magnetization, M, is given by

$$\frac{M(\infty) - M(t)}{M(\infty)} = \frac{3}{7}\exp(-\frac{3t}{T_1}) + \frac{4}{7}\exp(-\frac{10t}{T_1}) \tag{1}$$

and the spin-lattice relaxation time was uniquely determined by a fit of the data with the above formula.

III. RESULTS AND ANALYSIS

Fig. 1 indicates the evolution of the ^{55}Mn NQR spectrum for the $\pm 5/2 \leftrightarrow \pm 3/2$ transition at 4.2 K under pressure. With increasing pressure, the spectrum experiences a slight shift towards a higher frequency side, reflecting an increase of the electric field gradient (EFG). The full-width at half-maximum in a series of NQR spectra does not change appreciably, which means that the pressure-induced distribution in EFG can be ignored. The nuclear spin-lattice relaxation time T_1 was not measured above 120 K in order to avoid a melting of the transmitting liquid medium at high temperatures which increases the pressure effectively . The T dependence of $1/T_1T$, which is plotted with the semi-logarithmic scale in temperature (Fig. 2), shows the possible existence of two regimes marked by a kink (arrows). Hereafter, we will refer to this cross-over temperature as a characteristic spin fluctuation temperature, $T_{\rm sf}$, in the pressure-induced paramagnetic phase. $T_{\rm sf}$ increases from 30 K with increasing pressure. Below $T_{\rm sf}$, $1/T_1T$ becomes weakly temperature dependent. The spin-lattice relaxation rate, $1/T_1$, is in general related to the transverse component of the imaginary part in the dynamical magnetic susceptibility $Im\chi^{\perp}(q,\omega)$ such as

$$1/T_1 = \gamma_{\rm N}^2 N_0^{-2} T \sum_q A^2(q) \frac{Im\chi^{\perp}(q,\omega_0)}{\omega_0} \tag{2}$$

where $\gamma_{\rm N}$ is the gyromagnetic ratio of the nuclei studied, N the number of atoms, $A(q)$ the hyperfine coupling constant, and ω_0 the NMR frequency ($\perp$ represents the transverse component of the generalized susceptibility). In the case of nearly antiferromagnetic metals, the q dependence of the low-energy spin fluctuations are dominated by the components close of the AF vector **q=Q** and the main contribution will thus be such as $\chi^{\perp}(\mathbf{Q},\omega_{\mathbf{0}},\mathbf{T})$. Therefore Eq. (2) can be given by a simple form of

$$1/T_1T \propto A^2 \times Im\chi^{\perp}(\mathbf{Q},\omega_{\mathbf{0}},\mathbf{T}). \tag{3}$$

The pressure and thermal dependences of $1/T_1T$ thus allow us to probe corresponding dependences of AF spin fluctuation. In the framework of the SCR theory the dynamical susceptibility is given by

$$\chi^{\perp}(q,\omega) = \frac{\chi_0^{\perp}(q,\omega)}{1 - U\chi_0^{\perp}(q,\omega) + \lambda(q,\omega)} \tag{4}$$

where $\lambda(q,\omega)$ is the parameter to make the self-consistency equation valid, the non-interacting susceptibility $\chi_0^{\perp}(q,\omega)$ and U the Coulomb intra-site interaction. The denominator of Eq. (4) is equivalent to the Stoner factor which tends to zero at the criticality and P_{c}. Correspondingly the divergence of $Im\chi^{\perp}(q,\omega)$ means the divergence in the static staggered susceptibility and as a result, it is expected that $1/T_1T$ undergoes a divergence at P_{c} or it is largely enhanced approaching P_{c}. However, the experiment does not follow the above expectation. Rather, the development of AF spin fluctuations is saturated upon cooling and approaching the P_{c}. Namely, T_{sf} does not tend to zero, but seems to remain finite even at P_{c}. This may due to the first order nature of the transition from the AF state to paramagnetic at P_{c} induced by pressure. Above T_{sf}, the spin fluctuations are dominated by thermal fluctuations which are treated in terms of the SCR theory, whereas $1/T_1T$ approaches a constant value below T_{sf} characteristic for the Fermi liquid excitation. Fig. 3 indicates the pressure dependence of T_{sf} deduced from the present T_1 measurement. From the electrical resistivity measurements under pressure, the spin fluctuation temperature was defined as the cross-over temperature below which the thermal variation of $\rho(T)$ obeys the expected Fermi liquid T^2 law[1]. Remarkably, a fairly good agreement is obtained concerning the variation of both T_{sf} under pressure

$$\frac{\partial \ln T_{\mathrm{sf}}}{\partial P} = 59\mathrm{Mbar}^{-1} \tag{5}$$

With these experimental implications, it is possible that the development in spin fluctuation remains in short-range upon cooling.

IV. CONCLUSION

From the thermal variation of the nuclear spin lattice relaxation rate, $^{55}(1/T_1T)$ of ^{55}Mn above the critical pressure of 4 kbar, the spin fluctuation feature has been found to change below the temperature T_{sf}. At temperatures higher than T_{sf}, the nuclear relaxation behavior

is well described in terms of the self-consistent renormalized spin fluctuation theory for a nearly AF metal, whereas below T_{sf}, a deviation from that is significant and $1/T_1T$ exhibits a weak temperature variation. It is pointed out that T_{sf} coincides with the temperature below which a T^2 law in resistivity is valid as expected for the Fermi liquid state. We proposed that these features are understood from the view that the development in AF spin fluctuation remains in short-range associated with the singlet formation among Mn spins in each tetrahedron as suggested by the inelastic neutron experiments on $Y_{1-x}Sc_xMn_2$.

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FIGURES

FIG. 1. Pressure effect on the electric quadrupole frequency of the ^{55}Mn resonance $\pm 5/2 \leftrightarrow \pm 3/2$ at $T = 4.2$ K.

FIG. 2. Pressure effect on the thermal dependence of $1/T_1T$ of the ^{55}Mn nuclei in YMn_2.

FIG. 3. Comparaison of the pressure effect on the spin fluctuations temperature, $T_{\rm sf}$, in YMn_2 defined upon the present NQR experiment ($\bullet$) and by previous authors upon electrical resistivity measurements ($\circ$).